

ATTORNEYS AT LAW

NORMAN F. OBLON
(703) 413-3000
NOBLON@OBLON.COMSTEFAN U. KOSCHMIEDER, PH.D.
REGISTERED PATENT AGENT
(703) 412-6463
SKOSCHMIEDER@OBLON.COM

*Copy of ref. submitted
w/IDS on 3/10/04*

I hereby certify that this paper is being facsimile transmitted to the U.S. Patent and Trademark Office on the date shown below.

Date: 6/7/2004 Signature: *Stefan U. Koschmieder*
Attorney

To: Examiner: Price Telephone Number: 571-272-0644

Group Art Unit: 1621

Facsimile No.: 571-273-0644

From: Stefan U. Koschmieder Registration No.: 50,238

Telephone Number: 703-412-6463

Date: June 7, 2004

RE: U.S. Application Serial Number: 10/088,073

Filed: June 12, 2002

Attorney Docket Number: 220036US0X PCT

TOTAL NUMBER OF PAGES INCLUDING THIS PAGE: 3

COMMENTS

Please find attached a copy of the "76th Spring Meeting of Chemical Society of Japan, March 1999" reference (and English translation) provided with the IDS filed on March 10, 2004.

In the event that any fees are due, including any fees required under 37 CFR 1.136 for any necessary Extension of Time to make the filing of the attached documents timely, please charge the required fees to our Deposit Account No. 15-0030. Further, if these papers are not considered timely filed, then a petition is hereby made under 37 CFR 1.136 for the necessary extension of time.

1940 DUKE STREET ■ ALEXANDRIA, VIRGINIA 22314 ■ U.S.A.
TELEPHONE: 703-413-3000 ■ FACSIMILE: 703-413-2220 ■ WWW.OBLON.COM

Attached Sheet 1

Translation of the abstract presented in the 76th spring meeting of chemical society of Japan, 1999, Tokyo

2PB079 Intercalation of Anionic Fluorinated Surfactant into Layered Double Hydroxide
(Faculty of Engineering and Resource Science, Akita University) Shida Hiroaki; Fuda Kiyoshi;
Murakami Kenji; Matsunaga Toshiaki

[Introduction] Studies of intercalation of various molecules and ions into layered double hydroxides have been recently conducted aiming at new functionalities associated with regular array of guest molecular materials in the gallery. Along such direction, the intercalation of hydrocarbon surfactants in the LDH has attracted much attention in the field of chemistry. However, intercalation of fluorinated surfactants in whose structure the hydrogen atoms are substituted by fluorine ones has not been reported in literature concerning LDH materials. Fluorinated surfactants has fascinating characteristics such as thermal stability, water as well as oil repellency, ability of lowering the surface tension and so forth, which are considered to come from the unique nature of perfluoro chain of the surfactants. The purpose of this study is to clarify the possibility of intercalation of perfluorinated carboxylate type surfactant ($C_7F_{15}COONH_4^+$) into the gallery of Zn/Al-LDH [$x = Al^{3+}/(Zn^{2+} + Al^{3+}) = 0.33$], and to clarify the structural features of the intercalation compound obtained. Two preparation routes have been investigated: (1) ion exchange method via a LDH intercalated with the hydrocarbon type counterpart, $C_7H_{15}COOH^+$, (abbr. HC-LDH); (2) coprecipitation method by which the surfactant is directly inserted (abbr. FC-LDH).
[Experimental and Results] After preparing the HC-LDH, the ion exchange reaction proceeded with two-fold amount of the fluorinated surfactant for 1 day, and also for 7 days. On the other hand, the synthesis by the coprecipitation method was carried out using the surfactant of 1.2 fold in molar as much as the Al^{3+} ions embedded in the hydroxide sheet.

The XRD patterns for the intercalation compounds obtained are summarized in Fig. 1. The basal spacing of the HC-LDH was found to be 19.7, whereas those for 1-day and 7-day exchanged samples were found to be 28.5 and 48.0, respectively. On the other hand, that for the sample obtained by the coprecipitation method, FC-LDH-cop showed a series of the harmonic reflection lines extending to the 14th, corresponding to a basal spacing of 47.2, which suggested a goodness in regularity and crystallinity. This pattern was also found to be similar to that of 7-day exchanged sample. The intercalation of the surfactant was also examined by FT-IR and TG measurements. We speculate that the observed difference in structure despite of the same number of carbons in the alkyl chains of the surfactants is due to the difference in the cross section area between these two surfactants. A behavior associated with super repellency was also found for the surface of the FC-LDH, but not for the HC-LDH.

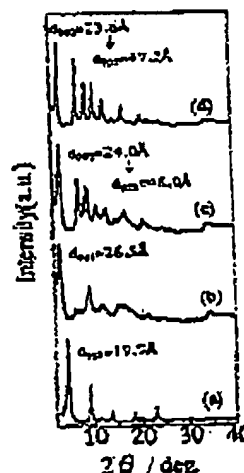


Fig. 1 XRD patterns for the composite products obtained by ion-exchange reaction and coprecipitation of the fluorinated surfactant into the LDH. (a) HC-LDH; (b) FC-LDH-ion1day; (c) FC-LDH-ion7days; (d) FC-LDH-cop.

BEST AVAILABLE COPY

Attached Seet 2

日本化学会第76春季年会(1999年3月於東京)講演予稿集

2 PB 079

フッ素系界面活性剤陰イオンの金属層状複水酸化物層間への挿入反応

(秋田大・工学資源) ○石田洋幸、市田源、村上賢治、松永利昭

Intercalation of Anionic Fluorinated Surfactant into Layered Double Hydroxide

(Faculty of Engineering and Resource Science, Akita University) Shida, Hiroaki; Fuda, Kiyoshi; Murakami, Kenji; Matsumaga, Toshiaki

[はじめに] 層状複水酸化物(LDH)層間へ様々な分子・イオン等を挿入する研究は、ゲスト物質の規則配列や複合化による新しい機能発現をねらい近年盛んに行われている。その中で、炭化水素系界面活性剤の層間挿入は、ミクロ2次元空間利用の可能性を広げる点で興味を持たれ LDH の化学の重要な関心事ともなっている。しかし、アルキル鎖のHをFに置き換えたフッ素系界面活性剤を LDH 層間に挿入した報告例は少ない。フッ素系界面活性剤は、パーフルオロアルキル基を持つため、熱的安定性や親水性や顕著な表面張力低下能など炭化水素系界面活性剤にはみられない魅力的な性質がある。そこで本研究では、 Zn/Al -LDH [$x = Al^{3+}/(Zn^{2+} + Al^{3+}) = 0.33$] 層間にカルボン酸塩型フッ素系界面活性剤($C_8F_{17}COO^-NH_4^+$)のインターカレーションの可能性と生成する層間化合物の構造と特徴を明らかにすることを目的とし、①炭化水素系界面活性剤($C_{18}H_{37}COO^-H$)挿入型 LDH(HC-LDH)を経由するイオン交換法(FC-LDH-ion)と②置換層間挿入する共沈法(FC-LDH-cop)について検討した。

[実験と結果] 予め HC-LDH を調製し、そのアニオン交換容量の2倍のフッ素系界面活性剤を用いて1日イオン交換をはかったのち、さらに7日間反応を進行させた。一方、共沈法による合成は、Al の1.2倍モルの界面活性剤を添加し、調製した。

生成挿入体の XRD 結果を Fig. 1 にまとめた。HC-LDH では層間間隔が 19.7Å であるのに対し、1日イオン交換体では 28.5Å、7日間イオン交換体では 48.0Å に変化した。一方、共沈法で得られた FC-LDH-cop は、47.2Å に対応する14次の回折が見られ、規則性・結晶性が良いことが分かった。このパターンは7日間イオン交換体に良く類似している。また、層間内へのフッ素系界面活性剤の取り込みは、FT-IR、TG 結果からも確認できた。各界面活性剤の炭素数が同一でもあるのに関わらず、挿入体の構造が違うのは、鎖の有効断面積の違いによるものと考えている。更にフッ素系界面活性剤挿入体表面は、炭化水素系界面活性剤挿入体では得られない超親水性の挙動を示した。

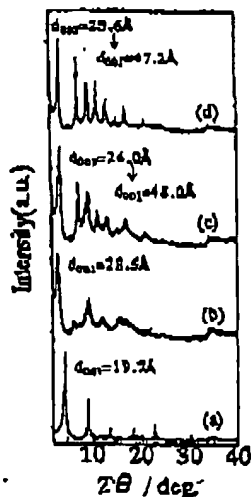


Fig. 1 XRD patterns for the composite products obtained by ion-exchange reaction and coprecipitation of the fluorinated surfactant into the LDH. (a) HC-LDH; (b) FC-LDH-1day; (c) FC-LDH-ion7days; (d) FC-LDH-cop.

2 PB

(関東学院)

Preparation and Dyea. (I)

MOBIL, Ic

1. 本研究で、トリメチルシリル基にフッ素のインターカレーションの検討を、2. クニピツ字)の2.5%レーシオンとウムクロリTEOS、ヒレピレンを、3. Ca^{2+} のイオンは12.5Åに粘土層間を、4. 液相中およびベクトルとに、5. ションにより2にその試験材料において、439nmの吸収スペクトルにより、6. モノマーの結晶より、7. マー状態で、8. ものと、9. 。

BEST AVAILABLE COPY